

Distinguishing different sources of organic aerosol in the particulate phase by application of deuterated precursors in chamber studies

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Atmospheric aerosol impacts the climate system by absorbing and scattering light (direct aerosol effect) and by serving as cloud condensation nuclei (indirect aerosol effect). Furthermore, high atmospheric aerosol loadings are detrimental for human health.

Organic Aerosol (OA) makes up a major fraction of total aerosol loading. OA can either be emitted directly from the earth surface (primary OA; POA) or can be formed by oxidation processes in the atmosphere (secondary OA; SOA). SOA contributes up to 80% of total OA on a global scale. However, the formation pathways of SOA remain not fully understood. Hence, systematic studies with different SOA precursors are essential for a better process understanding.

Thus far, one of the most challenging tasks has been the determination of the fraction of SOA formed from biogenic precursors (BSOA) such as plant emissions compared to the fraction of SOA formed from anthropogenic precursors (ASOA) such as gaseous emissions from road traffic and other combustion processes.

Here we use one isotopic labelled precursor to distinguish ASOA and BSOA in chamber studies. The Aerodyne Aerosol Mass Spectrometry (AMS) was applied on aerosol samples produced by oxidation of anthropogenic and biogenic precursors in the Jülich atmosphere simulation chamber SAPHIR. Real plant emissions from the new SAPHIR+ plant chamber (Hohaus et al, 2016) as biogenic precursors together with fully deuterated benzene (benzene-d₆) were introduced into the SAPHIR chamber. Oxidation of the precursors was driven by the OH radical which was photochemically produced from HONO decomposition by natural sunlight in the SAPHIR chamber (Rohrer et al, 2005).

In general the mass defect of D versus H is smaller than the resolving power of the AMS. Thus, peaks from deuterated fragments cannot be fully resolved from peaks of non deuterated fragments at most m/z. Nevertheless a subset of 20 peaks which are not overlapping with peaks from non-deuterated organic fragments could be identified. These peaks include oxygenized and nonoxygenized fragments as well as fragments with odd and even mass to charge ratios and can act as “marker” peaks since the ratio of these peaks to the total organic mass is constant throughout the experiment.

Figure 1 shows a typical time series of an experiment. After complete degradation of the biogenic precursors, benzene-d₆ was added to the chamber. The hereafter evolving SOA mass can be fully explained by the anthropogenic system.

Experiments were performed in different sequences of adding anthropogenic and biogenic precursors and under both low NO_x and high NO_x conditions in order to simulate realistic anthropogenic mixing scenarios and to test for different cross influences of the anthropogenic and biogenic precursors.

This study exhibits new insights in SOA chemistry and provides a tool to investigate interactions between anthropogenic and biogenic SOA precursors in different oxidation regimes.

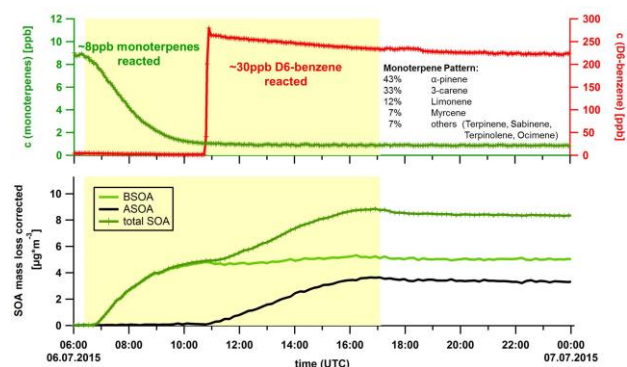


Figure 1. Time series of gaseous precursors (PTR-MS) and produced SOA (AMS) from a mixed SAPHIR experiment using *pinus sylvestris* emissions as BVOC. BVOC and benzene-d₆ concentrations were chosen to reach about the same OH reactivity. Yellow background indicates photo oxidation period.

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